

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventors : R. OKAMOTO et al.  
Serial No. : 10/576,227  
Filed : April 13, 2006  
For : **HIGH-STRENGTH STEEL SHEETS EXCELLENT IN  
HOLE-EXPANDABILITY AND DUCTILITY**  
Art Unit : 1733  
Conf. No. : 6918  
Examiner : Jie Yang

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

DECLARATION UNDER 37 C.F.R. §1.132

I, Eisaku Sakurada, hereby declare and state as follows:

1. I have been employed by Nippon Steel Corporation, Tokyo, Japan, to which the above patent application has been assigned, since April 1, 2007. My current position is a researcher in Nagoya Technical Research Department of Technical Development Bureau at Nippon Steel Corporation. I graduated from Tokyo Institute of Technology, with a Master's degree in department of metallurgy in 2007.

2. I have reviewed the specification and claims of the above-identified patent application. I have also reviewed the Office Actions mailed December 1, 2010 ("the December Office Action") and July 14, 2011 ("the July Office Action"), and references J.P. Patent Publication No. 2001-342543 to Tsutomu ("JP'543"), U.S. Patent No. 6,364,968 to Yasuhara et al. ("US'968") and U.S. Patent No. 5,470,529 to Nomura et al. ("US'529") cited therein.

3. I understand that the Examiner is of the opinion that:

- i) regarding the claimed composition, JP'543 (Abstract and claims 1-4 of JP'543) discloses compositions that overlap the "major composition ranges" of the instant invention, thus establishing a *prima facie* case of obviousness, and that although JP'543 does not disclose Al from 0.08-1.5 wt%, US'529 is applied for disclosing Al from 0.1-2.0 wt% in a similar steel alloy (December Office Action, at pp. 4-6);
- ii) regarding claimed equations (1)-(7) (claim 1) and Equations (1)-(6) and (8) (claim 9), the selection of proportions of elements Mg, O, S, Mn, Si, Al, C, Ti, and Nb requires no more than routine optimization (December Office Action, at pp. 6-7);
- iii) regarding claimed precipitates, JP'543 teaches the limitation of precipitates of MgO, MgS, and (Nb,Ti)N in the instant claims 1 and 9 (December Office Action, at p. 8); and
- iv) regarding the steel microstructure, JP'543 teaches a steel having a mainly ferrite microstructure and that US'968 teaches that microstructure is controllable making it obvious to adjust the microstructure of the JP'543 steel and arrive at the presently claimed steel of primarily bainite (claim 1) or ferrite + bainite (claim 9) (December Office Action, at pp. 4-5).

4. The presently claimed invention is directed to steel sheets having alloy compositions including both Al and Mg and achieving superior hole expandability, elongation, tensile strength, and chemical compatibility. Independent claim 1 is directed to steel sheets having a primarily bainite microstructure and independent claim 9 is directed to steel sheets having a primarily ferrite + bainite microstructure.

The inventors of the present invention discovered that Si and Al are very important elements for the structure control to secure ductility. However, Si may produce surface irregularities called Si-scale which are detrimental to product appearance, formation of chemical treatment films and adherence of paints. Thus, plentiful addition of Si is undesirable when chemical treatability is critical. Compatibility between ductility and chemical treatability can be obtained by substituting Al for Si. See, the present specification at page 15, lines 24-36. Conventionally, Al is added in an amount of 0.01 to 0.07% for deoxidation. The inventors also discovered that abundant addition of Al improves chemical compatibility and enhances ductility. However, addition of Al to more than 1.5% saturates the ductility enhancing effect and lowers chemical compatibility and deteriorates spot weldability. See, the present specification at page 9, lines 8-25.

The inventors of the present invention also discovered that the proportion of Si to Al needs to be limited according to equation (4):  $[Si\%]+2.2\times[Al\%]\geq0.35$  (4) for structure control, to secure ductility and in order to secure chemical compatibility and prevent surface irregularities, e.g., Si-scale, which are detrimental to formation of chemical treatment films and adherence of paints. Also, the total amount of both Si and Al must be limited to avoid excess ferrite and thereby secure the desired strength. Particularly when ductility is important, the combined content should preferably be not less than 0.9. See, the specification at page 15, line 37 through page 16, line 5.

Conventionally, sulfides are believed to cause deterioration of hole-expandability. However, the inventors discovered that in steel sheets having the amounts of added alloy elements within the presently recited ranges, hole-expandability and elongation can be remarkably improved by the precipitation of Mg-sulfides while impeding the precipitation of Mn-sulfides resulting in particle size refinement of precipitation of  $MgO$ ,  $MgS$  and  $(Nb,Ti)N$ . This particle size refinement can be achieved by controlling the relative amounts of alloy elements based on the presently recited equations.

For example, the inventors discovered that controlling the amounts of Mg, O and S according to equations 1-3 allows the precipitation of Mg-sulfides while impeding the precipitation of Mn-sulfides resulting in the combined precipitation of  $MgO$ ,  $MgS$  and  $(Nb,Ti)N$ , thus improving ductility and end-face properties.

Specifically, the inventors conceived that the amount of Mg relative to the amount of O must be controlled to produce Mg sulfides sufficient to improve hole expandability while retaining sufficient tensile strength. Mg must be greater than 80% of the amount of oxygen and that the amount of Mg should be controlled according to equation (1):  $[Mg\%]\geq([O\%]/16\times0.8)\times24$ . In equation (1), 16 and 24 correspond to the atomic mass of O and Mg, respectively. Thus, the right side of equation (1) represents the stoichiometric amount of Mg which combines with O to form the  $MgO$  precipitates.

Sulfur is essential in forming the required Mg-sulfides but also forms Mn-sulfides when present in large quantities causing deterioration of hole expandability. Based on this understanding, the inventors discovered that controlling the upper limit of S content with respect to Mg and O according to equation (2) inhibits MnS precipitation:  $[S\%]\leq([Mg\%]/24-[O\%]/16\times0.8+0.00012)\times32$ . In equation (2), the first term on the right side of the equation corresponds to the stoichiometric amount of Mg, the second term on the right side

corresponds to the stoichiometric amount of O which combines with Mg, the third term on the right side represents a factor in accordance with securing the inventive properties, and 32 represents S atomic mass.

The inventors discovered that constraining the proportion of S to Mn according to equation (3):  $[S\%] < 0.0075/[Mn\%]$  prevents Mn-sulfide precipitation based on the solubility product of Mn and S. If the S content is greater than the limit set by equation (3), a large quantity of Mn-sulfide precipitates and hole-expandability deteriorates. As the specification discloses, large quantities of Mn and S inhibit Mg-sulfide production and prevent sufficient improvement of hole-expandability. See, the specification as filed at page 14, lines 1-5.

Thus, controlling the amounts of Mg, O, S, and Mn according to equations (1) to (3) allows sufficient Mg-S precipitation based on the discovery that Mg-sulfides can be used for achieving uniform and fine precipitation of (Nb, Ti)N, which conventionally has not been recognized. The inventors also discovered that not less than  $3.0 \mu\text{m}$  of the combined precipitates of MgO, MgS and (Nb, Ti)N must be present; e.g., the steel sheet must contain composite precipitates of MgO, MgS and (Nb, Ti)N of not smaller than  $0.05 \mu\text{m}$  and not larger than  $3.0 \mu\text{m}$  in an amount of not less than  $5.0 \times 10^2$  per square millimeter and not more than  $1.0 \times 10^7$  per square millimeter.

Furthermore, the inventors discovered that limiting the contents of C, Mn, Ti, and Nb in steels primarily containing bainite (claim 1) is effective for securing ductility while maintaining strength and good hole-expandability by improving the end-face properties of punched holes by Mg-precipitates. See, specification at page 16, lines 31-36. The proportion of C and Ti controlled by equation (5) ensures hole expandability and the proportion of C to Mn is controlled by equation (6) balances strength and ductility. In order to secure strength in excess of  $980 \text{ N/mm}^2$  it is necessary to control the proportion of C, Mn, Ti and Nb in accordance with equation (7).

In order to secure an adequate amount of ferrite in the steels primarily containing ferrite/bainite steel (claim 9) effective for the enhancement of ductility, C, Si, Mn and Al contents must satisfy equation (8):  $-100 \leq -300[\text{C}\%] + 105[\text{Si}\%] - 95[\text{Mn}\%] + 233[\text{Al}\%]$ . If the value of equation (8) is smaller than -100, ductility deteriorates because an adequate amount of ferrite is not obtained and the percentage of the second phase increases. See, the specification at page 20, lines 1-8.

Thus, the present invention provides special kinds of steel sheets having superior hole expandability, elongation, tensile strength, and chemical compatibility made possible by a combination of alloy content ranges, equation constraints on the relative amounts of alloy elements, as well as the size distribution of composite precipitates. In particular, the present invention allows addition of a large amount of Al for improving chemical compatibility without sacrificing tensile strength, hole expandability, and elongation. The recited equations are not general formulae, but are critical constraints on the *relative amounts* of added elements that are not disclosed by the prior art.

Test data disclosed in the instant application support the criticality of controlling the alloy contents, the relative amounts of alloy elements in accordance with the claimed equations, and the type and size distribution of composite precipitates in achieving improved elongation and hole expandability at the high tensile strengths of the inventive steel. For example, in Example 1 corresponding to bainite steels, comparative steels a, b, c, d, e, f and g disclosed in Tables 1-4 and shown in Table A and Fig. A, below, and in Example 3 corresponding to ferrite/bainite steels, comparative steels a, b, c, d, e, f and g disclosed in Tables 11-14 and shown in Table B and Fig. B, below, correspond to steels in which either at least the content of an alloy element or the distribution of composite precipitates is outside the claimed range (open triangles), or the composition and the distribution of composite precipitates are both within the claimed ranges, but at least one of the equations is not satisfied (open squares).

Table A. Example 1, Comparative Bainite Steels. Excerpt from Tables 1- 4, Steels corresponding to alloys that do not satisfy the limitations of claim 1.

Comparative Steel	element outside range	equation not satisfied	Precipitates not in claimed range	Tensile Strength N/mm <sup>2</sup>	% Elongation	% Hole Expandability
a	C 0.210%	(5)		1067	7	10
b	Mn 3.6%	(3)		1178	5	51
c	O 0.006%	(1), (2)		1001	16	45
d		(4)		1009	6	68
e	S .0100%	(2), (3)		1014	14	43
f	Mg 0.0003%	(1)	3x10 <sup>2</sup>	1000	17	39
g		(7)		896	19	44

Fig. A

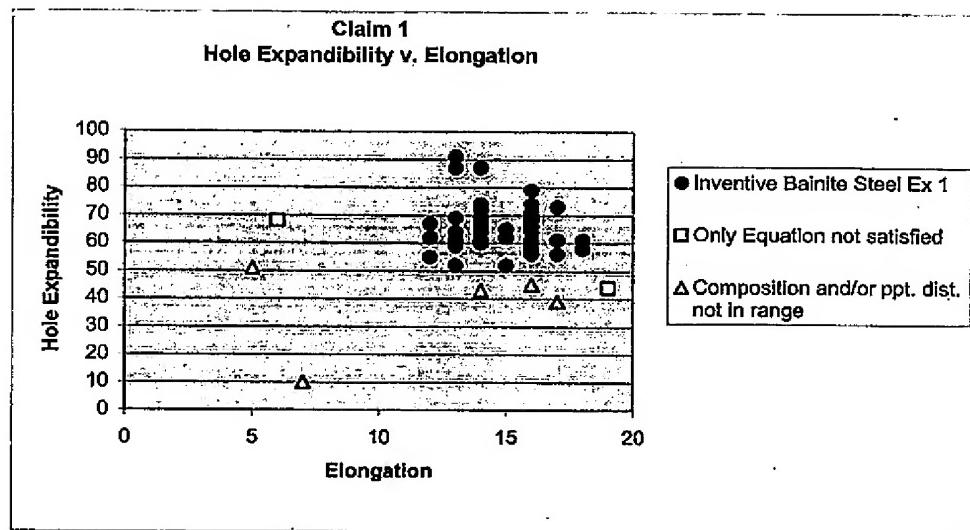
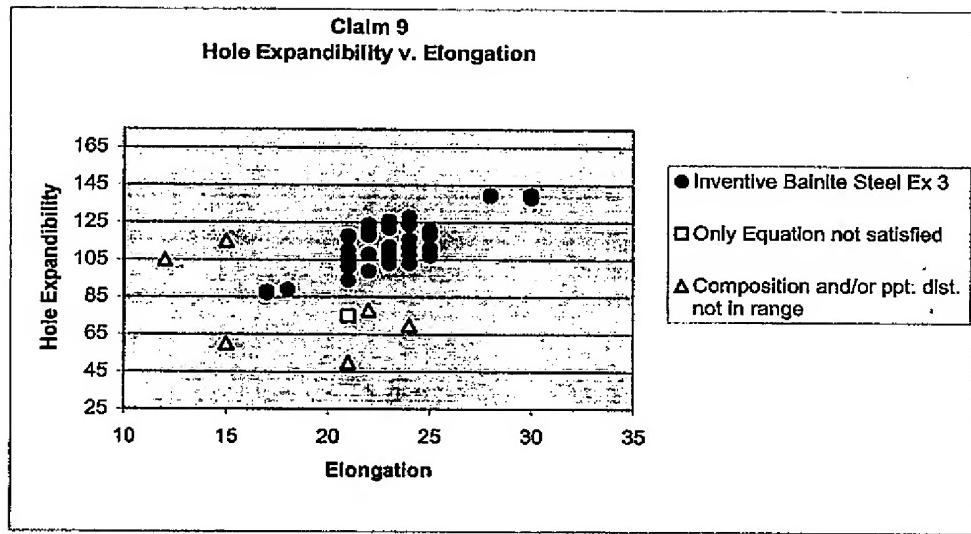


Table B. Example 3, Comparative Ferrite/Bainite steels. Excerpt of Tables 11-14, Steels corresponding to alloys that do not satisfy the limitations of claim 9.

Comparative Steel	Element outside range	Equation not satisfied	Precipitates	Tensile Strength N/mm <sup>2</sup>	% Elongation	% Hole Expandability
a	C 0.210%			795	15	60
b	Mn 3.6%	(3), (8)		859	12	105
c	O 0.006%	(1), (2)		850	21	50
d		(4), (8)		782	15	115
e	S .0100%	(2), (3)		749	24	70
f	Mg 0.0003%	(1)	5.0×10 <sup>2</sup> /mm <sup>2</sup>	788	22	78
g			1.0×10 <sup>7</sup> /mm <sup>2</sup>	812	21	75

Fig. B



Figs. A and B show that in these comparative steels, at least one of elongation or hole expandability is inadequate, which can be seen clearly as the open squares and triangles representing these steels fall into regions of lower elongation and/or hole expandability. Comparative steels having the content of an alloy element outside the claimed range, *i.e.*, steels a, b, c, e and f, are shown as open triangles, and comparative steels meeting all of the claim limitations except that they fail to satisfy at least one of the claimed equations, *i.e.*, steels d and g in Example 1 and steel d in Example 3, are shown by open squares. Figure A demonstrates that for bainite steels, at least steels not satisfying equations (4) and (7) would not achieve both high hole expandability and high elongation, and Figure B demonstrates that for ferrite + bainite steels, at least steels not satisfying equation (4) would not achieve both high hole expandability and high elongation. Thus, the combination of alloy content ranges, equation constraints on the relative amounts of alloy elements, as well as the size distribution of composite precipitates are critical for the production of the inventive steels having excellent hole expandability and elongation at high tensile strength.

5. A comparison of the claimed invention and the cited references is shown in Table C, below.

**Table C**

	Instant Claim 1	Instant Claim 9	JP'543	US'529	US'968
Al	0.18-1.5	0.18-1.5	<b><u>0.002-0.07</u></b>	0.1-2.0	$\leq 0.150$
Mg	0.0006-0.01	0.0006-0.01	0.0005-0.01	<u>not disclosed</u>	<u>not disclosed</u>
Eqs. (1)-(8)	Eqs. (1)-(7)	Eqs. (1)-(4) and (8)	<u>not disclosed</u>	<u>not disclosed</u>	<u>not disclosed</u>
Precipitate size distribution	MgO, MgS, (Nb,Ti)N	MgO, MgS, (Nb,Ti)N	<u>MgS not disclosed</u>	<u>Mg not disclosed</u>	<u>Mg not disclosed</u>
Microstructure	Mainly Bainite	Bainite + Ferrite	<u>Mainly Ferrite</u>	Mainly Ferrite or Ferrite + Bainite	Mainly Bainite

From Table C it can be seen that these references not only do not teach or suggest all of the features of the steel sheets of the present invention even if they are combined, but also teach certain features that are either incompatible with the steel sheets of the present invention or incompatible with each other, discussed below.

For example, none of the references teach or suggest controlling the relative amounts of alloy elements according to the claimed equations or the size distribution and composition of the composite precipitates. As discussed above, both of these features are critical for the production of the inventive steels having excellent hole expandability and elongation at high tensile strength.

The steels of both US'543 and US'968 contain low Al, and teach that increasing Al in their steels has detrimental effects. For example, JP'543, which concerns an invention which utilizes Mg for suppressing the formation of coarse cracks and improving hole expansibility, discloses that increasing Al to above 0.07% impairs the effects Mg (*see, e.g., JP'543 at ¶ [0024]*). US'968 discloses that Al deteriorates surface properties and reduces strength, and is limited to 0.15 or less, but for stability of mechanical properties, Al is preferably limited to 0.010 to 0.080% (*see, e.g., US'968, col. 7, lines 13-18*).

On the other hand, the steels of both US'529 and US'968 contain no Mg, which is critical in the steels of the present invention and US'543. Furthermore, the steels of US'543, US'529, and US'968 all have microstructures different from each other.

I wish to point out that it was not predictable that combining US'529, US'968, and US'543 in the manner proposed by the Examiner would provide the steel sheets of the present invention, *i.e.*, having superior hole expandability, elongation, tensile strength, and chemical compatibility. Figs. C-E below show overlays of hole expandability, elongation, tensile strength of the steel sheets of the present invention, US'529, US'968, and US'543. Fig. C below shows a comparison of hole expandability vs. tensile strength of the inventive bainite steels of claim 1, *i.e.*, Example 1 in Tables 1-4 in the specification at pages 27-33; the inventive bainite+ferrite steels of claim 9, *i.e.*, Example 3 in Tables 11-15 in the specification at pages 41-46; US'543, *i.e.*, the inventive steels listed in Tables 2 and 4 at ¶¶ [0047] and [0049]; US'968, *i.e.*, the inventive steels listed in Tables 3 and 5; and US'529, *i.e.*, the inventive steels listed in Tables 3-1 and 3-2. Fig. D shows a comparison of elongation vs. tensile strength, and Fig. E shows a comparison of the hole expandability vs. elongation of the inventive steels and those of the cited references. A linear fit to each data set is also plotted.

In Figs. C and E, the hole expandability data of US'968 are obtained by converting the disclosed hole-expandability ratio such that the data are properly comparable. Specifically, US'968 reports the results of the hole expansion test as "hole-expandability ratio." Although US'968 does not specifically disclose the definition of hole-expandability ratio, US'968 discloses at column 1, line 63 through column 2, line 2, by reference to JP patent publication no. 62-196336 ("JP'336"), which disclose hole expanding ratio as  $d/d_0$  on page 4 and reports values around 1.5 (See, Tables 1 and 2 on page 4), that its hole-expandability ratio is defined as  $d/d_0 \times 100$ . Since each of the present invention, JP'543 and US'529 define percent hole-expandability as  $\lambda = (d - d_0)/d_0 \times 100$ , I have converted the US'968 hole expanding ratio values to hole-expandability for comparison.

Fig. C

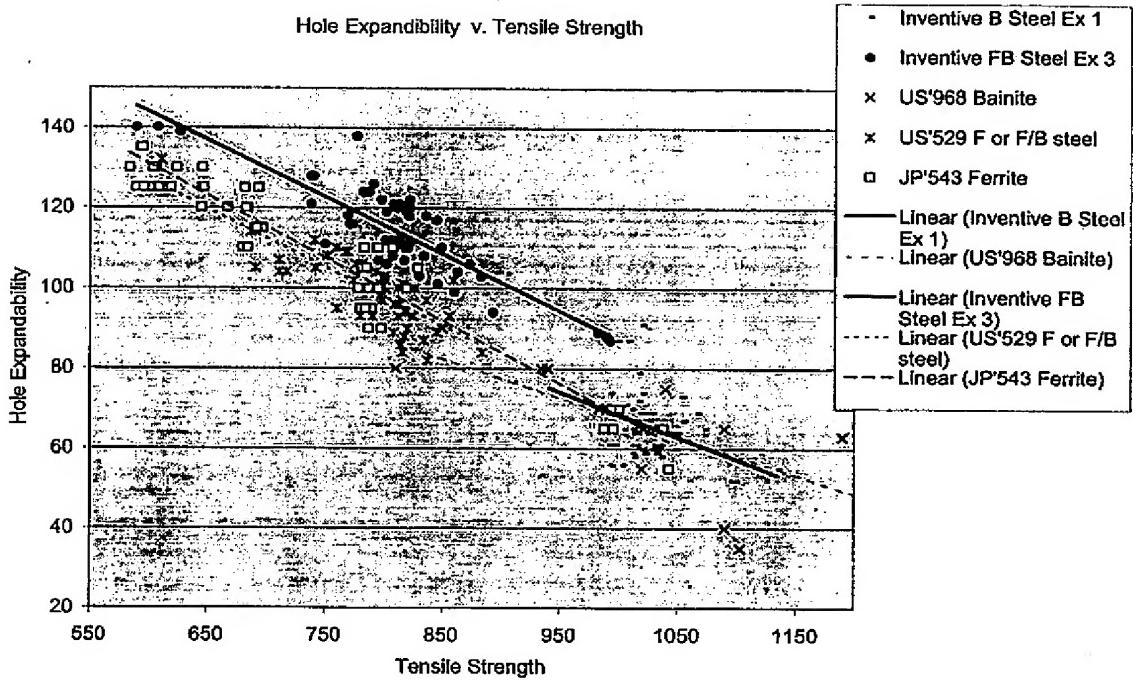


Fig. D .

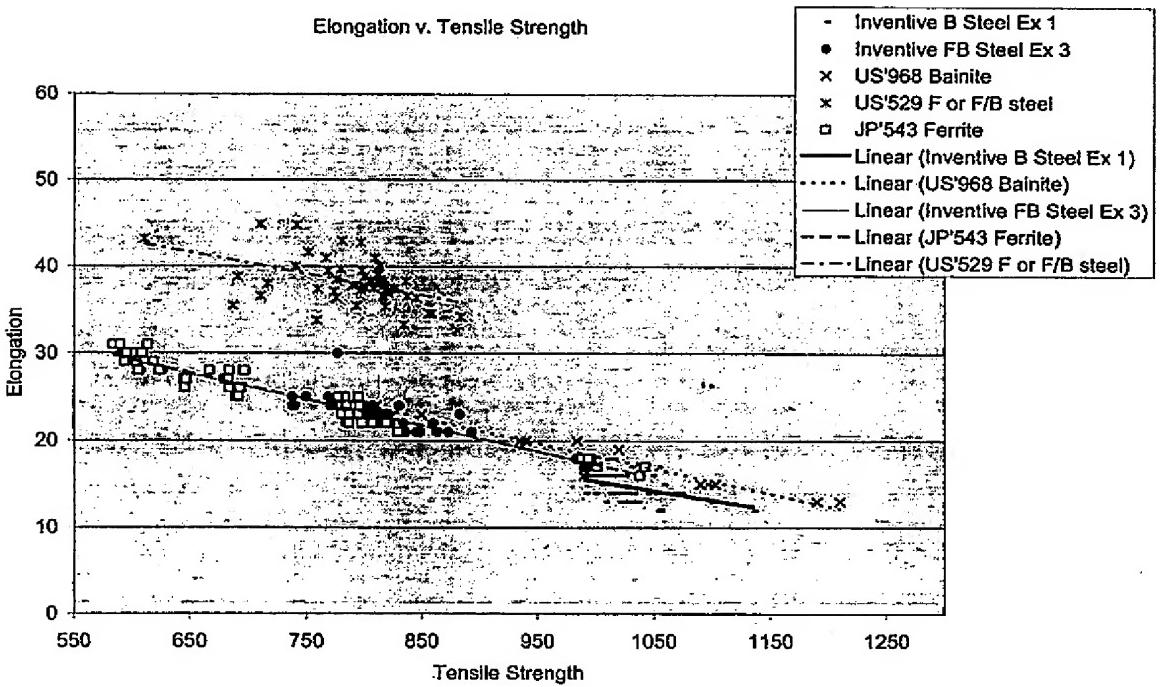
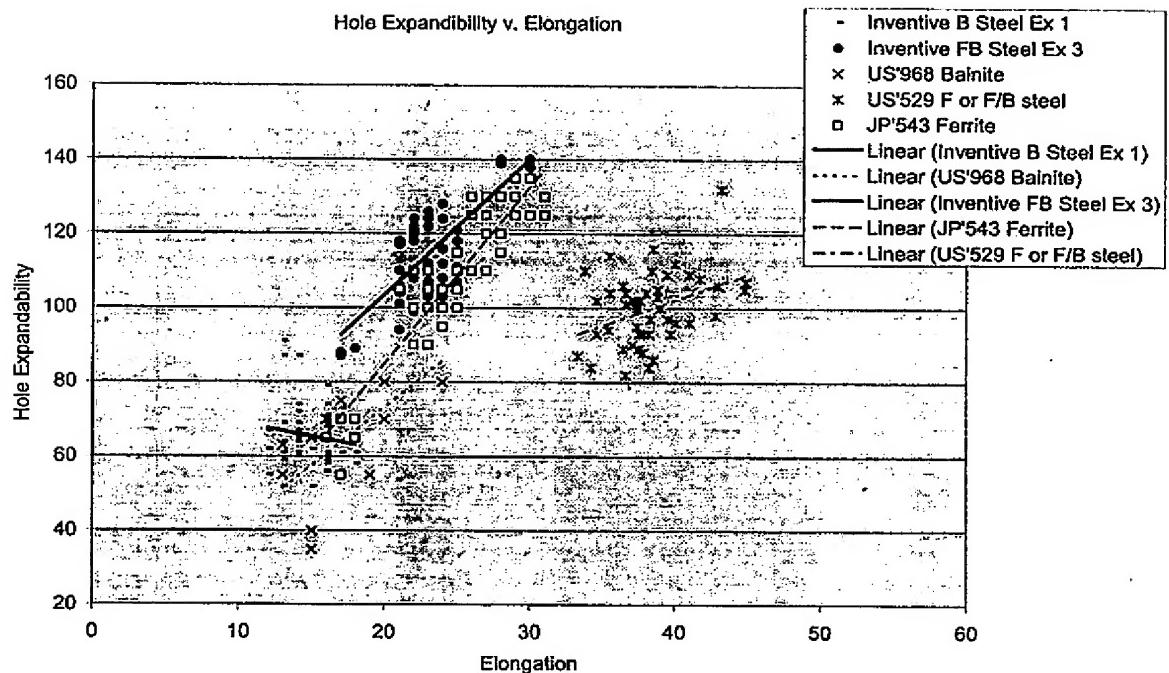


Fig. E



It can be seen from Fig. C that the hole expandability of the ferrite+bainite steel sheet of the present invention (claim 9) is clearly higher than those of US'543 and US'529. If extrapolated to higher tensile strength according to the linear fit, the hole expandability of the ferrite+bainite steel sheet of the present invention is expected to be higher than those US'968. Similarly, it can be seen from Fig. E that the hole expandability of the ferrite+bainite steel sheet of the present invention (claim 9) are higher than those of US'543 and US'968. If extrapolated to higher elongation according to the linear fit, the hole expandability of the ferrite+bainite steel of the present invention (claim 9) is expected to be higher than those US'529.

On the other hand, Fig. C shows that the hole expandability of the bainite steel sheet of the present invention (claim 1) is comparable to those of US'543 and US'968, even though the bainite steel sheets of the present invention contains a high Al content and better chemical compatibility. In addition, the bainite steel sheet of the present invention (claim 1) has significantly higher tensile strength than those of US'529.

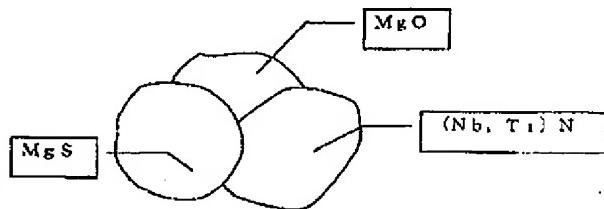
These comparisons demonstrate that even if the references are combined as proposed by the Examiner, it cannot be predicted that the steel sheets of the present invention can be

achieved.

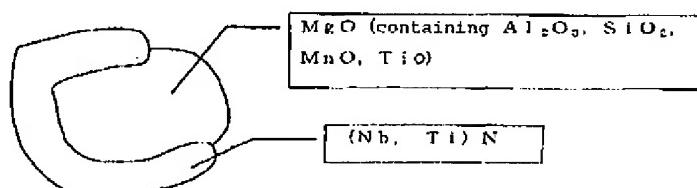
Figs. C, D and E also show that the properties such as tensile strength, hole expandability, and elongation of the steel sheets of US'543, US'529, and US'968 are very different from each other, as revealed by different regions in the plots. It cannot be predicted where the properties of a steel sheet formed by combining these references would fall, e.g., whether the properties would fall in the region of US'543, or the region of US'529, or the region of US'968; or a region anywhere in-between or even entirely away.

6. Regarding the structure of the steel sheet, I wish to point out that only JP'543 discloses steel sheet containing Mg. However, JP'543 teaches MgO inclusions, or combined precipitates such as  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ ,  $\text{MnO}$  and  $\text{Ti}_2\text{O}_3$ , or combined precipitates surrounded by  $(\text{Nb}, \text{Ti})\text{N}$ , as shown in the following Fig. F, all of which are quite different from the precipitates of the present invention. More specifically, JP'543 describes that MgO is preferable with one or more complex oxides such as  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ ,  $\text{MnO}$  and  $\text{Ti}_2\text{O}_3$ . See, JP'543 at ¶ [0027]. Further, Mg and  $\text{MgAl}_2\text{O}_4$  mainly have an effect of form of fine void by means of neighboring precipitation of  $(\text{Nb}, \text{Ti})\text{N}$  neighboring those complex oxides, and it is considered MgO and  $\text{MgAl}_2\text{O}_4$  contribute as nuclei for uniform distributed precipitation. See, JP'543 at ¶ [0028]. Fig. F, below, shows a comparison of the present inventive combined precipitates and an example of the inclusions described in JP'543.

Fig. F  
Present inventive combined precipitates



Inclusions disclosed by JP'543



Thus, none of the references disclose steel sheets having the precipitates of the present invention. As explained above, controlling the type and size distribution of the precipitates is critical to achieve the excellent hole-expandability and elongation in the steel sheets of the present invention. Therefore, the steel sheets of the present invention cannot be obtained based on JP'543, US'529, and US'968.

7. Based on the above discussion, it is my opinion that the cited references would not have led one of ordinary skill in the art to the present invention.

\* \* \* \* \*

I hereby declare that all statements made herein of my own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the patent or any reexamination certificate issued.

Respectfully submitted,

January 23, 2012  
Date

Eisaku Sakurada  
Eisaku Sakurada